

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Appl. No. : 10/572,935
Applicants : Guenther HAMBITZER *et al.*
Filed : March 22, 2006
TC/A.U. : 1795
Examiner : Adam A. Arciero

Docket No. : 2945-176
Customer No. : 06449
Confirmation No. : 4983

DECLARATON OF CHRISTIANE RIPP PURSUANT TO 37 C.F.R. § 1.132

I, Christiane RIPP, hereby declare and state as follows:

1. I studied chemistry at the University of Karlsruhe and graduated with a diploma in chemistry ("Dipl.-Chem.") in February 1995. From March 1995 to April 1999, I did post-graduate studies and thesis work. I received a Ph.D ("Dr. rer. nat"), from the University of Witten/ Herdecke. The scientific experimental work for my thesis was performed at Fraunhofer Institute for Chemical Technology at Pfinztal/Germany. From July 2000 to July 2002, I was employed as chemical scientist at Fortu Bat Batterien GmbH in Pfinztal/Germany. Since then, I have worked as a freelance chemist and taken care of my two children.

2. Both during my Ph.D work and during my employment at Fortu Bat, I participated in experimental work with electrochemical battery cells. I am a co-inventor of international patent application PCT/DE 04/02105 and of the corresponding U.S. patent application to which this Declaration refers ("present application"). My thesis work was part of a cooperation between the Fraunhofer Institute for Chemical Technology and Fortu Bat. Therefore, I was also involved in the development work of Fortu Bat on which international

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patent application PCT/DE 00/00177 (PCT Pub. No. WO00/44061) ("Hambitzer '061")¹ and the corresponding US patent 6,709,789 are based. Furthermore, although not an inventor of international patent application PCT/DE 00/01801 (PCT Pub. No. WO00/79631) ("Hambitzer '631")² and the corresponding U.S. patent 6,730,441, I am familiar with the development work of Fortu Bat on which they were based.

3. As a freelance chemist, I work *inter alia* for Fortu Research GmbH, Karlsruhe, and Fortu Intellectual Property AG, Switzerland. The CEO of Fortu Research GmbH, Karlsruhe is Dr. Günther Hambitzer, co-inventor of the above identified patents and patent applications. As a consultant of these companies, I have full access to all documentation concerning the experimental work from which above-identified applications and patents resulted and also to all scientific results that were generated after the initial filing of the patent applications.

4. I have carefully studied the file of the present application, including the final office action of September 3, 2010, the applicant's reply of November 10, 2010, and the advisory action of December 14, 2010, as well as the prior art cited by the Examiner of the USPTO.

5. The advisory action of December 14, 2010, asserts that Hambitzer '631 describes or suggests embodiments of a battery cell having no separator acting as a barrier layer impermeable to active metal. The Examiner's basis for the assertion is that Hambitzer '631 discloses that "[a] separator layer can be applied directly to this foamlike substrate" and that the cells of Examples 1 and 2 of Hambitzer '631 do not explicitly list a separator as a component.

¹ As in the final office action of September 3, 2010, cites to the text of Hambitzer '061 refer to the corresponding U.S. patent 6,709,789.

² As in the final office action of September 3, 2010, cites to the text of Hambitzer '631 refer to the corresponding U.S. patent 6,730,441.

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6. Based on my familiarity with the experimental work on which Hambitzer '631 was based and the documentation thereof, battery cells having no separator acting as a barrier layer impermeable to active metal were not in consideration during the experimental work on which Hambitzer '631 was based. In fact, the documentation of the experimental work on which Hambitzer '631 was based does not include experiments carried out on battery cells having no separator acting as a barrier layer impermeable to active metal.

7. Based on my familiarity with the experimental work on which the present application was based and the documentation thereof, experimental work on battery cells having no separator acting as a barrier layer impermeable to active metal did not begin until the year 2002, well after completion of the experimental work on which Hambitzer '631 was based. Further, the locally delimited short-circuit behavior of the battery cells containing an electrolyte based on SO_2 , a porous insulator arranged and formed such that it is possible for active mass deposited on the negative electrode to grow during charging of the cell to the surface of the positive electrode, and no separator acting as a barrier layer impermeable to active metal were completely unexpected at that time.

8. Accordingly, based on my familiarity with the experimental work on which the present application was based and the documentation thereof and my familiarity with the experimental work on which Hambitzer '631 was based and the documentation thereof, it is my opinion that Hambitzer '631 in no way describes or suggests embodiments of a battery cell having no separator acting as a barrier layer impermeable to active metal.

9. Moreover, based on my experience in the field, it is my opinion that one of ordinary skill in the art would understand Hambitzer '631 to only describe and suggest battery

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cells having a separator acting as a barrier layer impermeable to active metal. First, Hambitzer '631 is directed to the avoidance of short-circuits, which are safety problems. Hambitzer '631 at col. 4, lines 42-55.

10. Second, contrary to the Examiner's assertion, Hambitzer '631 explicitly discloses that the electrochemical cell of Example 1 has a separator formed as part of the negative electrode by applying aluminum oxide to the nickel foam of the negative electrode. Hambitzer '631 at col. 7, lines 23-26. Hambitzer '631 also discloses that the cell of Example 2 was prepared similar to the cell of Example 1 and no changes to the negative electrode or separator were disclosed. *Id.* at col. 7, lines 47-54.

11. Third, Example 3 does not explicitly disclose a separator, but one of ordinary skill in the art would understand that this is only because a conventional separator is used and not the special type of separator formed as part of the negative electrode as in Examples 1 and 2. Examples 1 and 2 use a "nickel foam" to which a separator can be applied directly while Example 3 uses a "precision-expanded nickel foil." *Compare* Hambitzer '631 at col. 7, lines 23-26 and 47-54 *with Id.* at col. 8, lines 5-6. One of ordinary skill in the art would understand that, in Example 3 of Hambitzer '631, a conventional separator, which is not applied directly to the negative electrode made of nickel foil, would be used and that no further description mention of the conventional separator would be necessary. *See Id.* at col. 6, line 62-col. 7, line 2.

12. Fourth, one of ordinary skill in the art would not understand the statement in Hambitzer '631 that "[a] separator layer can be applied directly this foamlike substrate" to mean that a separator layer is not required as asserted by the Examiner. In contrast, one of ordinary in the art would understand the statement to mean that either a separator layer applied directly to a

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metallic foamlike substrate of the negative electrode could be used or a separator layer that is not applied directly to a metallic foamlike substrate of the negative electrode (*i.e.*, a conventional separator layer) could be used. *See Id.* at col. 6, line 62-col. 7, line 2.

13. Accordingly, based on my experience in the field, it is my opinion that one of ordinary skill in the art would understand Hambitzer '631 to only describe and suggest battery cells having a separator acting as a barrier layer impermeable to active metal.

14. From the above, it is clear that, at the time the invention was made, the inventors did not have an embodiment without a separator acting as a barrier layer impermeable to active metal. It is my opinion that one of ordinary skill in this art, when reading the Hambitzer '631 disclosure, would recognize that the inventors did not contemplate such an embodiment, and that it therefore would not be obvious to such a person to include a porous insulator layer that was not impermeable to active metal.

15. The final office action of September 3, 2010, and advisory action of December 14, 2010, assert that the structure of the present application and the structure disclosed in Hambitzer '631 are the same.

16. Based on my familiarity with the experimental work on which Hambitzer '631 was based and the documentation thereof, the battery cells of Hambitzer '631 does not have a structure that permits active metal deposited on the negative electrode to grow through a porous insulator layer to the surface of the positive electrode only at locally delimited points. In contrast, the battery cells of Hambitzer '631 include a barrier layer impermeable to active metal that prevents active metal deposited on the negative electrode to grow to the surface the positive electrode. Further, if active metal deposited on the negative electrode were to break through the

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barrier layer of Hambitzer '631 to the positive electrode, the breakthrough would not be locally limited, and the contact of the active metal to the positive electrode (*i.e.*, short-circuit) would cause an uncontrolled reaction sequence called "thermal runaway."

17. Based on my familiarity with the experimental work on which the present application was based and the documentation thereof, the battery cell of the present application has a structure that permits active metal to grow through the porous insulator layer but only at locally delimited points.

18. Although the specific values and design features of the porous insulator layer cannot be specified with general validity, whether a particular material is suitable for use as a porous insulator layer in a particular battery cell may be determined by using the experimental setup illustrated in Fig. 3 of the current application. A test material is suitable for use as a porous insulator layer in a particular battery cell in accordance with the present application if active metal penetrates the test material only at locally delimited points.

19. Based on my familiarity with the experimental work on which Hambitzer '631 was based and the documentation thereof, if the separator that acts as a barrier layer impermeable to active metal in a particular battery cell in accordance with the disclosure of Hambitzer '631 were tested using the experimental setup illustrated in Fig. 3 of the current application, active metal would not penetrate the separator.

20. Accordingly, based on my familiarity with the experimental work on which the present application was based and the documentation thereof and my familiarity with the

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experimental work on which Hambitzer '631 was based and the documentation thereof, it is my opinion that the battery cells of the present application have a structure different than the battery cells disclosed by Hambitzer '631.

21. Based on my experience in the field, one of ordinary skill in the art would understand that the pores of a porous insulator layer is arranged and formed such that it is possible for active mass to grow the pores of the insulator have a size larger than the pores of a separator acting as a barrier layer and impermeable to active mass.

22. Based on my experience in the field, one of ordinary skill in the art would understand that the reduction of organic oligomers has no applicability to an inorganic electrolyte based on SO₂.

23. Regarding claim 19, which refers to a battery cell containing an electrolyte which is based on SO₂, the electrolyte is, in a battery cell, the means that provides the ionic transport between the electrodes. In an SO₂-based electrolyte, the SO₂ forms the "solvent" for the ions of the conductive salt. The mobility of the ions, which allows the charge transport between the electrodes, is ensured by the SO₂.

24. This is a fundamental difference as compared to normal lithium-ion-cells in which the conducting salt is dissolved in an organic solvent such as ethylencarbonate. The electrolyte of a cell is decisive with respect to any reactions of the materials contained in the electrodes, including unwanted reactions which may disturb the performance of the cell. The materials of the electrodes (generally solid) are only in contact with the electrolyte (generally liquid) and consequently can only react with its constituents. Therefore, the reactions that may take place in

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an organic SO₂-based electrolyte are completely different from those that may take place with the same electrode materials in an organic electrolyte.

25. For the specific case of an SO₂-based electrolyte, the inventors have found that the passivation of electrodes may be attributed to a reaction of surface molecules of the electrode with water (*e.g.*, from ambient humidity). The reaction products comprise a hydroxide. From this surprising finding, the inventors derived the teaching of claim 19, namely that, in cells having an SO₂-based electrolyte, it is advantageous for the electrode surface to be essentially free of hydroxide ions. In contrast to the inventors, the Examiner of the US PTO does not consider this finding surprising and instead considers it obvious over a combination of Hambitzer '061 and US patent 6,596,440 Gavelin *et al.* ("Gavelin").

26. Despite claim 19 being rejected over the combination Hambitzer '061 in view of Gavelin, in the Advisory Action of December 14, 2010, the Examiner responds to arguments regarding claim 19 by citing to Hambitzer '631. Although it is unclear exactly on which Hambitzer reference the Examiner is actually relying, based on my experience in the field, no combination of the cited references would have led one of ordinary skill in the art to the invention recited in claim 19.

27. Hambitzer '631 is a patent originating from our research group. It refers to a SO₂-based battery cell and is mainly concerned with an aspect that has no relationship to the present invention, namely the total amount of SO₂ contained in the cell as compared to the electrical charge of the active metal which can be theoretically accumulated in the positive electrode. In the Advisory Action of December 14, 2010, the Examiner states that Hambitzer '631 (i) "discloses the same structure of battery as the present disclosure", (ii) "discloses that the positive

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electrode does not have to be LiCoO_2 and that it can be carbon or a metal halide and wherein the negative electrode can be sodium, calcium or zinc (col. 1, lines 15-22)” and (iii) “discloses electrodes (those not containing LiCoO_2) which are free of hydroxide ions.” Thus, the Examiner is apparently taking the position that the other electrode materials described by Hambitzer ‘631 must (inherently) be essentially free of hydroxide ions. Based on my experience in the field, this assumption is not correct.

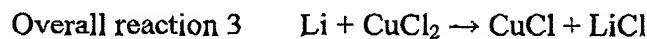
28. Regarding sodium, calcium and zinc, the Examiner evidently refers to column 1, lines 20 to 25 and claim 14 of Hambitzer ‘631. However, Hambitzer ‘631 suggests these elements as active metals forming the positive ion of the conductive salt and not as forming an insertion electrode containing active metal. Based on my experience in the field, one of ordinary skill in the art would not understand the invention of claim 19 to refer to active metals forming the positive ion of the conductive salt. Rather, one of ordinary skill in the art would understand amended claim 19 to make perfectly clear that the condition regarding the freedom of the electrode surface from hydroxide ions refers to an insertion electrode which contains the active metal in its interior. Consequently, based on my experience in the field, one of ordinary skill in the art would not understand the positive metal ions of Hambitzer ‘631 to correspond to the feature recited in claim 19.

29. With respect to metal halides, for example CuCl_2 , the Examiner evidently refers to column 1, lines 49 to 52 of Hambitzer ‘631. As stated therein, metal halides have been proposed in the Handbook of Batteries (1994) as a material for the positive electrode making a simple electrode reaction with the active metal. These electrode materials are not insertion

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electrodes, and based on my experience in the field, one of ordinary skill in the art would not understand them to correspond to the feature recited in claim 19.

30. For example, the copper(II)chloride / Lithium system, as mentioned in Hambitzer '631, is a system with two independent electrode reactions. The negative electrode consists of metallic lithium, the positive electrode comprises copper(II)chloride as the active mass. At the negative electrode, during discharging of the cell, metallic lithium is oxidized to lithium ions (reaction 1). At the positive electrode, at the same time, copper(II)chloride is reduced to copper(I)chloride (reaction 2).



There is no active metal in the interior of the copper chloride (nor the negative electrode) and no moving of the positive lithium ions into the interior of the positive electrode during charging or discharging of the cell, like in a lithium system with an insertion electrode. Copper chloride is not able to take up lithium ions; it is not an insertion electrode. Accordingly, based on my experience, one of ordinary skill in the art would not understand the disclosure of Hambitzer '631 to correspond to the feature recited in claim 19.

31. Carbon is mentioned in Hambitzer '631 as a possible material of the positive electrode. Here, it functions as a current collector and catalyst material for cells with liquid cathode materials like sulphur dioxide (SO₂), thionyl chloride (SO₂Cl) or sulfuryl chloride (SO₂Cl₂). Such liquid cathodes can be used with the SO₂-based electrolyte described in Hambitzer'631. These cells use a lithium anode and a porous carbon electrode to catalyze the

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reaction of the liquid active cathode material. The carbon doesn't take active part in the electrochemical reactions. It serves as a catalyst and current collector. There is no moving of the positive lithium ions into and out of the interior of the catalyst during charging or discharging of the cell, like in a lithium system with an insertion electrode. Carbon, as a catalyst at the positive electrode, is not able to take up lithium ions. In this case, it is not an insertion electrode, and, based on my experience, one of ordinary skill in the art would not understand the carbon to correspond to the feature recited in claim 19.

32. The Examiner seems to have difficulty accepting that the results of the experiments disclosed in the patent specification (with reference to Figs. 5 to 8) confirm the presence of hydroxide ions on the surface of LiCoO_2 electrodes and the superior results (regarding internal resistance and electrical capacity of the cell) achieved with electrodes which are essentially free of hydroxide. See Advisory Action. However, based on my experience in the field, it is unquestionable that one of ordinary skill in the art would understand the results of the experiments disclosed in the patent specification (with reference to Figs. 5 to 8) to confirm the superior results achieved with electrodes that are essentially free of hydroxide. Furthermore, based on my experience in the field, one of ordinary skill in the art, at least at the time of the invention, would have understood the superior results to be unexpected.

33. According to Applicants' Specification, Figures 5 and 6 show cyclic voltammograms which have been obtained using an electrode not optimized according to the present invention and, using an electrode optimized according to the present invention, respectively. The electrode material is lithium cobalt oxide. Lithium cobalt oxide is an insertion electrode material and also mentioned by Hambitzer '631 as possible electrode material.

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34. The present invention teaches to optimize the electrode before using the electrochemical cell:

According to the second main aspect, the OH^- ions are removed from the electrodes using a cleaning agent which contains a first cleaning component reacting with OH^- ions. It is contacted with the electrode in such a manner that OH^- ions bonded thereto are removed from the electrode surface by reaction with the first cleaning component, and components of the cleaning agent or reaction products which may interfere with the function of the cell are removed from the electrode.

Applicants' Specification at p. 12, lines 18. Neither Hambitzer '631 nor Hambitzer '061 disclose a method to clean the insertion electrode or to remove the OH^- -ions of the surface of the electrode. There is no hint, that OH^- -ions of the surface will degrade the properties, like the shift of the peak potential or an increase of the internal resistance of the electrode.

35. The not optimized electrode material of the experiments of Figures 5 to 8 is not cleaned and the OH^- -ions of the surface are not removed. This "not optimized material" is comparable to the material of Hambitzer '631. In contrast, the optimized electrode material is cleaned as described in the present invention, and the OH^- -ions of the surface are removed. Therefore, the results shown in Figs. 5-8 clearly show improvement over known cells, for example over the known cell of Hambitzer WO'631 and are unexpected, because, in the context of the present invention, it has been established that the presence of chemically bound water may result in passivation of the electrodes, so that the electrode processes are inhibited.

36. With respect to Gavelin, the Advisory Action states that it is aimed at solving a similar problem (passivation layers) for achieving similar results (improved battery performance). Thus, far I agree with the examiner. Certainly electrode passivation is a known

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problem in the battery technology and a person skilled in the art would aim to avoid or remove same.

37. However, the invention refers to a particular type of passivation in a particular cell, namely passivation caused by the presence of hydroxide ions on the surface of an insertion electrode (adapted for taking up positive metal ions of a conducting salt into its interior during charging of the cell) while this electrode is in contact with an electrolyte which is based on SO₂. The cell design of claim 19 requires the knowledge described in paragraph 25 above, namely that in a SO₂-based electrolyte, electrode passivation is caused by a reaction of surface molecules of the electrode with water, the reaction products comprising a hydroxide. Gavelin provides no information which may be useful in this context.

38. Gavelin refers to battery cells having an organic electrolyte (as in conventional lithium-ion batteries), whereas the invention refers to a cell with an inorganic SO₂-based electrolyte. Gavelin refers to a type of passivation which occurs in organic electrolytes but can not occur in an inorganic electrolyte on the basis of SO₂. The oligomers causing the problem in the case of Gavelin are simply not present in an SO₂-based electrolyte. On the other hand, the passivation problems in an inorganic SO₂-based electrolyte are completely different from those in an organic electrolyte environment. See paragraphs 23 and 24 above.

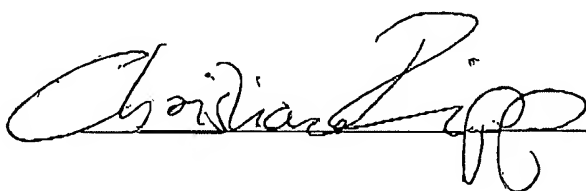
39. For these reasons, a person skilled in the art would not have considered Gavelin when trying to solve any problems in a SO₂-cell. However, even if she or he referred to Gavelin, nothing useful could be found therein regarding problems of passivation in a SO₂-cell.

40. I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that

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these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Date:

2/2/2011A handwritten signature in black ink, appearing to read "Christian Zipp", written over a horizontal line.